

Atmospheric delivery of anthropogenic bioavailable iron from mineral dust to the ocean

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Atmospheric deposition of anthropogenic soluble iron (Fe) to the ocean has been suggested to modulate primary ocean productivity and thus indirectly affect the climate. A key process contributing to anthropogenic sources of soluble Fe is associated with air pollution, which acidifies Fe-containing mineral aerosols during their transport and leads to Fe transformation from insoluble to soluble forms. However, there is large uncertainty in our estimate of this anthropogenic soluble Fe. Here, we interactively combined laboratory kinetic experiments with global aerosol modeling to more accurately quantify anthropogenic soluble Fe due to air pollution. We firstly examined Fe dissolution kinetics of African dust samples at acidic pH values with and without ionic species commonly found in aerosol water (i.e., sulfate and oxalate). We then constructed a new empirical scheme for Fe release from mineral dust due to inorganic and organic anions in aerosol water, by using acidity as a master variable. We implemented this new scheme and applied an updated mineralogical emission database in a global atmospheric chemistry transport model to estimate the atmospheric concentration and deposition flux of soluble Fe under preindustrial and modern conditions. Our improved model successfully captured the inverse relationship of Fe solubility and total Fe loading measured over the North Atlantic Ocean. However, our modeled Fe solubility was significantly lower than that deduced from observations over the South Atlantic east downwind from the Patagonian dust source regions. Our modeled Fe solubility for dry deposition over the Atlantic is in good agreement the measurement, while that for wet deposition is significantly lower than the measurement. Our model results suggest that human activities contribute to about half of the soluble Fe supply to a significant portion of the oceans in the Northern Hemisphere, while their contribution to oceans in the high latitude remains highly

uncertain due to limited understanding of dust blown off the coasts of Alaska, Iceland and the Patagonia desert.